

Coexistence of Magnetic Fluctuations and Superconductivity in the Pnictide High Temperature Superconductor $\text{SmFeAsO}_{1-x}\text{F}_x$ Measured by Muon Spin Rotation

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Muon spin rotation experiments were performed on the pnictide high temperature superconductor $\text{SmFeAsO}_{1-x}\text{F}_x$ with $x = 0.18$ and 0.3 . We observed an unusual enhancement of slow spin fluctuations in the vicinity of the superconducting transition which suggests that the spin fluctuations contribute to the formation of an unconventional superconducting state. An estimate of the in-plane penetration depth $\lambda_{ab}(0) = 190(5)$ nm was obtained, which confirms that the pnictide superconductors obey an Uemura-style relationship between T_c and $\lambda_{ab}(0)^{-2}$.

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The recent discovery of high temperature superconductivity (HTSC) in the layered tetragonal pnictide compound $\text{RFeAsO}_{1-x}\text{F}_x$ ($R = \text{La, Nd, Pr, Gd, and Sm}$) with critical temperatures T_c above 50 K came as a considerable surprise [1–3]. This is the first family of non-copper-oxide-based layered superconductors with T_c exceeding 40 K and raises the expectation that even higher T_c values can be achieved. This discovery also gives rise to the speculation that a common pairing mechanism is responsible for HTSC in cuprates and pnictides.

At first glance, the pnictides appear rather different from the cuprates. Band structure calculations suggest that they are multiband superconductors with up to five FeAs-related bands crossing the Fermi level [4–6] as opposed to the cuprates which have only one relevant $\text{Cu}(3d_{x^2-y^2})\text{O}$ band. A more complex exchange interaction is suggested since, besides the indirect $\text{Fe}(3d)\text{-As}(4p)$ hybridization, a sizable direct $\text{Fe}(3d)\text{-Fe}(3d)$ overlap has been predicted [5,6] as well as significant frustration [7]. Furthermore, electron doping gives the highest T_c here, compared to hole doping for the cuprates.

Nevertheless, there are also some striking similarities, such as HTSC emerging on doping away from a magnetically ordered parent compound [8]. Neutron measurements on undoped LaFeAsO have revealed commensurate spin-density wave (SDW) order of the Fe moments below $T_N = 135$ K with amplitude $0.35\mu_B$ [9], a result confirmed by muon spin rotation (μSR) and Mössbauer [10]. Resistivity measurements exhibit an anomaly near T_{SDW} in LaFeAsO and SmFeAsO which has been tracked as a function of F doping. These measurements suggest that the magnetic order is rapidly suppressed upon doping and that the maxi-

mum T_c is achieved just as static magnetism disappears [8]. Recent neutron measurements on superconducting (SC) samples confirm this conjecture since they could not detect any magnetic order [9]. Thus an important issue is whether weak, slowly fluctuating or disordered magnetism persists in these superconductors.

In this Letter we report a μSR study which provides new insight into the magnetic properties of this superconductor. Two polycrystalline samples with nominal compositions of $x = 0.18$ and 0.3 were synthesized by conventional solid state reaction methods [2,8]. All (the main) peaks in standard powder x-ray diffraction patterns could be indexed to the tetragonal ZrCuSiAs -type structure for $x = 0.18$ ($x = 0.3$), as previously reported [2,8]. Measurements of the dc resistivity and magnetization were made to determine T_c (ΔT_c) = 45(3) and 45(4) K for $x = 0.18$ and 0.3 corresponding to the midpoint (10%–90% width) of the resistive and the diamagnetic transitions.

The μSR experiments were performed at the EMU, MuSR, and ARGUS instruments of the ISIS facility, Rutherford Appleton Laboratory, UK, which provides pulsed beams of 100% spin polarized muons. μSR measures the time evolution of the spin polarization of the implanted muon ensemble using the time-resolved asymmetry $A(t)$ of muon decay positrons. The technique [11,12] is well suited to studies of magnetic and SC materials as it allows a microscopic determination of the internal field distribution and gives direct access to the volume fractions of SC and magnetic phases [12].

Figure 1(a) shows representative spectra of the zero-field (ZF) μSR measurements at three different temperatures for $x = 0.18$. The relatively fast relaxation of $A(t)$, which

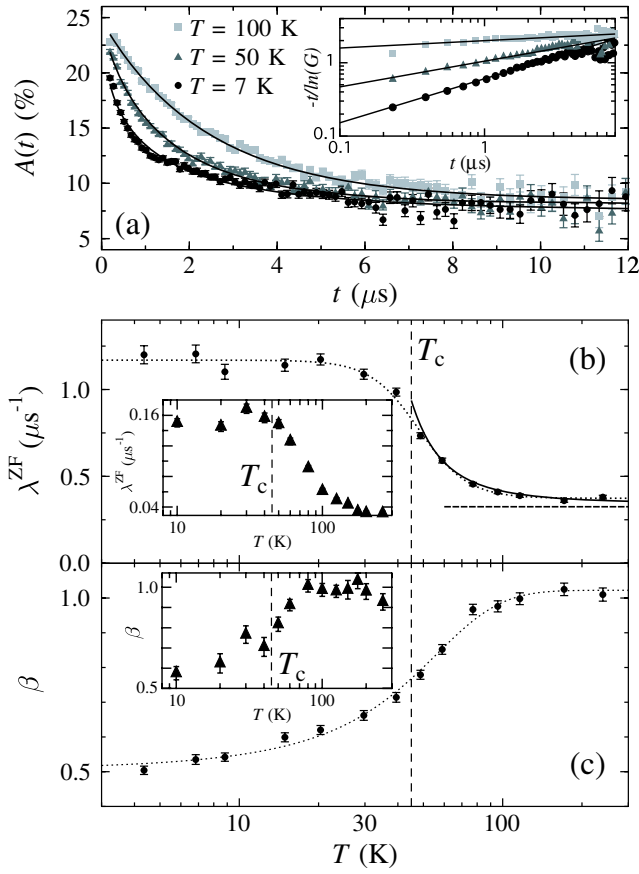


FIG. 1 (color online). (a) Example ZF- μ SR spectra for $x = 0.18$. Inset: log-log plot of the polarization function $G(t)$ [13]. (b) Temperature evolution of λ^{ZF} for $x = 0.18$. Inset: Corresponding data for $x = 0.3$. (c) Shape parameter β for $x = 0.18$. Inset: Corresponding data for $x = 0.3$. In (b) the solid line is the sum of a temperature-independent component (horizontal dashed line) and an activated component with an activation energy of 13 meV. Dotted lines are a guide to the eye.

persists even at 200 K, provides clear evidence for the presence of sizable electronic magnetic moments. We find that these spectra are well described with a single stretched exponential relaxation function of the form $A(t) = A(0)G(t)$ where the spin polarization function is $G(t) = \exp[-(\lambda^{\text{ZF}}t)^\beta]$. This is illustrated in the inset of Fig. 1(a) which shows that the data follow straight lines on a log-log plot of $-t/\ln(G(t))$ versus t [13]. The temperature dependences of the relaxation rate λ^{ZF} and the exponent β are shown in Figs. 1(b) and 1(c), respectively. Above 100 K the relaxation is exponential with $\beta \approx 1$ and λ^{ZF} is only weakly temperature dependent. Below 100 K the value of λ^{ZF} exhibits a significant increase followed by a saturation below 30 K with a low temperature value of $\lambda^{\text{ZF}} \approx 1.2 \mu\text{s}^{-1}$. At the same time β decreases continuously towards $\beta \approx 0.5$. Notably, the biggest changes occur in the vicinity of the SC transition at $T_c = 45(3)$ K, as shown by the vertical dashed line in Figs. 1(b) and 1(c). The inset of Fig. 1(b) shows corre-

sponding data for the $x = 0.3$ sample where λ^{ZF} is reduced but exhibits a similar temperature dependence.

In order to distinguish between static and dynamic contributions, a longitudinal field (LF) scan was performed at 60 K (Fig. 2). We observe an abrupt transition in the longitudinal relaxation rate λ^{LF} at around 40 G but subsequent increases in LF produce no further significant change. This unusual behavior cannot be accounted for by a simple decoupling model for purely static or dynamic spins (dashed line in Fig. 2) which would predict $\lambda^{\text{LF}} \propto B^{-2}$ above some critical field (corresponding either to the internal field in the static case or ν/γ_μ in the dynamic case, where ν is the fluctuation rate).

Two potential sources for the muon relaxation are the lanthanide moments in the SmO layers and magnetic fluctuations originating from spin correlations in the FeAs layers. The strong doping dependence of λ^{ZF} [Fig. 1(b)] demonstrates clearly that the observed magnetism cannot be explained solely in terms of weakly coupled Sm moments. The data for $T > T_c$ naturally separates into two components: T -independent and T -dependent, as indicated in Fig. 1(b). The latter component has an activation energy in the range of 10–20 meV, a typical scale for lanthanide moment fluctuations [14]. The amplitude corresponds to the nonquenched component seen in LF and we ascribe this component to Sm moments which are fluctuating rapidly due to crystal field excitations [14]. The temperature-independent component which is quenched in 40 G at 60 K can be identified with low energy spin fluctuations associated with the FeAs layer.

Although the high temperature relaxation is simple exponential and can therefore be associated with a single, dominant fluctuation rate, cooling through T_c results in a reduction in β , signifying a range of fluctuation rates and/or local field amplitudes. This indicates that the spin dynamics at low temperature becomes substantially more complex. Below T_c , the activated behavior ceases and λ^{ZF} saturates, demonstrating that an additional relaxation channel becomes dominant. Although relaxation with $\beta =$

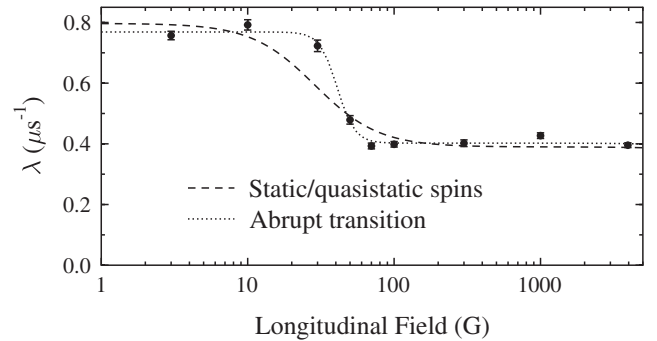


FIG. 2. Field dependence of λ^{LF} at 60 K for the $x = 0.18$ sample. The dashed line shows a simulation assuming decoupling from a simple static or dynamic local field distribution (see text). The dotted line is a guide to the eye and illustrates the more abrupt transition which is actually observed.

0.5 can be suggestive of glassy behavior or static disorder, it is unlikely to be the case here. Since it is known that the Sm moments order antiferromagnetically (AFM) below $\sim 3\text{--}4\text{ K}$ [15] (and see below), a static disordered Sm state at higher temperature is precluded. While it is possible that static Fe moments could develop in this temperature regime, the lack of change in λ_{ZF} in the region below 30 K where β is changing continuously makes this interpretation unlikely. We can also discard any interpretation of our data which involves a small fraction of ordered spins, since the observed relaxation corresponds to the behavior of the overwhelming bulk of the sample across the entire temperature range; thus the role of any purported minority phase is not directly probed in these experiments.

Our data therefore suggest that the onset of SC is accompanied or slightly preceded by the enhancement of slow spin fluctuations which originate (at least partially) in the FeAs layers. This raises the question of whether this coincidence is accidental or signifies that the spin fluctuations are playing an active role in the SC pairing mechanism. The former scenario is not supported by the similar temperature dependences of the spin fluctuations for the $x = 0.18$ and $x = 0.3$ samples (despite the different absolute values). Furthermore, the latter scenario agrees with the finding that T_c for the Sm compounds are almost twice those of the La ones, whose spin fluctuations appear to be considerably weaker or possibly even absent [16,17]. It is also supported by theoretical calculations which show that spin fluctuations emerging in the proximity of an AFM or SDW state can mediate or at least significantly enhance a singlet SC state [18,19]. Notably, the spin fluctuations enhance only unconventional order parameters whereas they suppress conventional ones.

One might even be tempted to speculate about an unconventional SC state with spin triplet Cooper pairs, similar to (U, Th)Be₁₃, UPt₃ [20], and Sr₂RuO₄ [21] where the breaking of time-reversal symmetry yields spontaneous supercurrents that create internal magnetic fields below T_c . However, these magnetic fields should be static rather than dynamic. Also the Sm moment would have to play an important role in this unconventional SC state since no corresponding increase in the ZF relaxation rate has been observed in the related La compound [16,17].

Certainly, our observations call for further investigations to clarify the role of slow spin fluctuations in the SC pairing mechanism and to explore whether the enhanced spin fluctuations in the Sm compound as compared to the La one are brought about by the coupling to the lanthanide moments or rather by the related structural changes [1,2].

Unconventional superconductivity is also supported by our transverse field (TF) μ SR measurements which yield a low temperature value of the in-plane magnetic penetration depth $\lambda_{ab}(0)$ that falls close to the so-called Uemura line of the cuprate HTSC [22]. The TF- μ SR spectra for $B_{\text{app}} = 100\text{ G}$ were well described with a sum of two Gaussian functions using the form

$$A(t) = \sum_{i=1,2} A_i(0) \cos(\gamma_\mu B_i t) \exp\left[-\left(\frac{\sigma_i^{\text{TF}}}{2} t\right)^2\right], \quad (1)$$

where A_i , B_i , and σ_i^{TF} correspond to the amplitude, the local magnetic field at the muon site, and the relaxation rate, respectively. The second weakly damped component reflects the small background from muons not stopping in the sample. The first, dominant component is due to the sample, and its temperature dependence is shown in Fig. 3. A sharp rise of σ^{TF} [Fig. 3(a)] is seen below T_c which exceeds that which would be expected from the ZF data; this additional contribution reflects the formation of the vortex lattice. This interpretation is confirmed by an observed diamagnetic shift of $\sim 13\text{ G}$ [plotted in Fig. 3(b)] which also occurs at T_c . The additional steep rise of σ^{TF} below 4 K [see inset of Fig. 3(a)] likely represents additional local field broadening due to the ordering of the Sm moments.

Notably, on cooling towards T_c there is a gradual onset of a diamagnetic shift already at about 70 K, which is detailed in the inset of Fig. 3(b). As was already noted above, this could be an indication for a precursor SC state with a transition temperature higher than the bulk T_c or the onset of SC fluctuations above T_c . However, at present we cannot rule out the possibility that the slowing down of the spin fluctuations leads to this reduction of the local field. In any case, the sharp onset of the diamagnetic shift at T_c and the corresponding increase in σ^{TF} allow us to provide an estimate of the in-plane magnetic penetration depth. Allowing for an additional root-exponential damping in

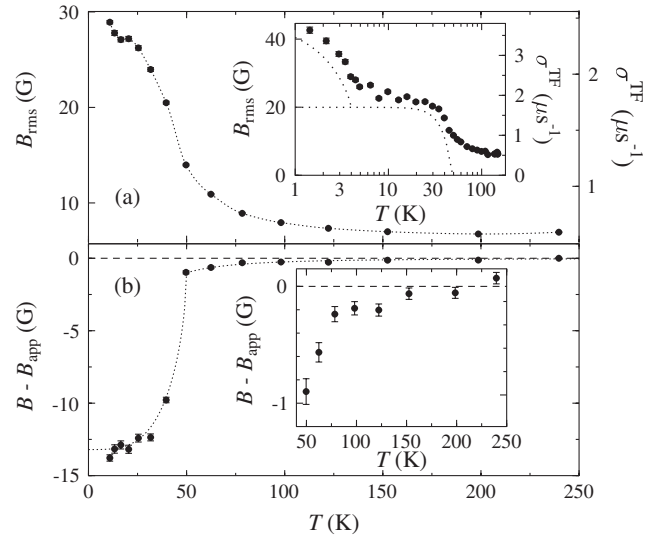


FIG. 3. (a) The main component of σ^{TF} (ARGUS spectrometer). The inset shows additional data below 4 K (MuSR spectrometer), which reveals a steep increase of σ^{TF} , likely due to the ordering of the Sm moments. The dashed lines illustrate the respective contributions from the SC vortex lattice and the Sm ordering. (b) The diamagnetic shift due to the development of the SC state. Inset: A detail of the behavior above T_c . Dotted lines are guides to the eye.

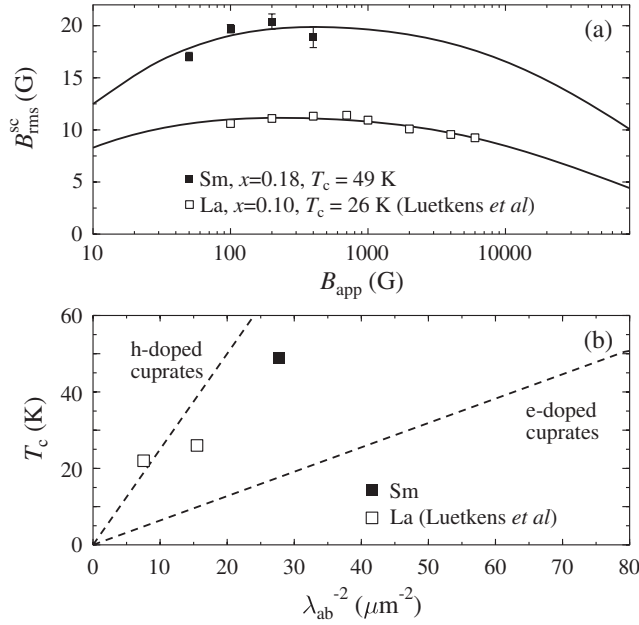


FIG. 4. (a) The field dependence of the SC vortex contribution to the rms linewidth of our data. The fitted field dependence is given by a powder averaged version of the model in Ref. [23]. Data for the La compound from Ref. [16] are shown for comparison [24,25]. (b) The Uemura plot for the pnictide superconductors measured to date by μ SR. The trends observed for the cuprates are shown as dashed lines for comparison.

the dominant term of (1) to take account of the contribution to the relaxation from magnetic fluctuations (which is known from the ZF measurements), the SC vortex contribution to the total linewidth $B_{\text{rms}}^{\text{SC}}$ is obtained. This is plotted in Fig. 4(a) against applied field at 10 K and from this data we derive $\lambda_{ab} = 190(5)$ nm. This estimate is derived from fitting to the numerical results of a recent detailed Ginzburg-Landau vortex lattice calculation [23], taking a polycrystalline average for our powder sample in the high anisotropy limit, under the assumption that the length scales λ and ξ diverge following $1/\cos\theta$ as the field orientation approaches the plane at $\theta = 0$.

Since the estimate is made at $0.2T_c$, it should provide a good account of $\lambda_{ab}(0)$, assuming a two-fluid type of saturating temperature dependence. Note that we are unable to establish whether any additional gap-node related linear term might be present at low temperatures due to the extra relaxation contribution below 4 K. Our value of λ_{ab} is shorter than those found for $\text{LaFeAsO}_{1-x}\text{F}_x$ by Luetkens *et al.* [254(2) nm for $x = 0.1$ and 364(8) nm for $x = 0.07$ [16,24]], reflecting the higher T_c of our compound and hence larger superfluid stiffness (proportional to λ_{ab}^{-2}). Figure 4(b) shows the Uemura plot [22] for the pnictide superconductors measured to date by μ SR. Apparently the overall trend lies closer to that of the hole-doped than the electron-doped cuprates.

In conclusion, the μ SR results on polycrystalline $\text{SmFeAsO}_{1-x}\text{F}_x$ with $x = 0.18$ and 0.3 provide clear evi-

dence for the coexistence and interplay of superconductivity and dynamic magnetic correlations. The magnetic correlations exhibit a complex temperature dependence, and a significant contribution of magnetic fluctuations to the enhanced T_c is suggested. From TF measurements we obtained an estimate of the in-plane magnetic penetration depth of $\lambda_{ab} = 190(5)$ nm, which comes rather close to the Uemura line of the hole-doped cuprates.

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Note added.—Since submission of this Letter we learned of Ref. [26] which interprets the ZF- μ SR of $\text{SmFeAsO}_{0.85}$ with two exponential components and attributes the relaxation solely to weakly coupled Sm moments. Such an interpretation is inconsistent with our relaxation data and its LF and doping dependences.

- [1] Y. Kamihara *et al.*, J. Am. Chem. Soc. **130**, 3296 (2008).
- [2] X.H. Chen *et al.*, Nature (London) **453**, 761 (2008).
- [3] Z.A. Ren *et al.*, arXiv:0803.4283.
- [4] S. Lebegue, Phys. Rev. B **75**, 035110 (2007).
- [5] K. Haule *et al.*, Phys. Rev. Lett. **100**, 226402 (2008).
- [6] G. Xu *et al.*, Europhys. Lett. **82**, 67002 (2008).
- [7] T. Yildirim, Phys. Rev. Lett. **101**, 057010 (2008).
- [8] R.H. Liu *et al.*, arXiv:0804.2105.
- [9] C. de la Cruz *et al.*, Nature (London) **453**, 899 (2008).
- [10] H.-H. Klauss *et al.*, Phys. Rev. Lett. **101**, 077005 (2008).
- [11] S.J. Blundell, Contemp. Phys. **40**, 175 (1999).
- [12] See, for example, A. Schenck, *Muon Spin Rotation: Principles and Applications in Solid State Physics* (Adam Hilger, Bristol, 1986).
- [13] R.H. Heffner *et al.*, Phys. Rev. B **63**, 094408 (2001).
- [14] R.I. Bewley *et al.* Phys. Rev. B **60**, 12286 (1999).
- [15] L. Ding *et al.*, Phys. Rev. B **77**, 180510(R) (2008).
- [16] H. Luetkens *et al.*, Phys. Rev. Lett. **101**, 097009 (2008).
- [17] J.P. Carlo *et al.*, arXiv:0805.2186.
- [18] M. Kato *et al.*, J. Phys. Soc. Jpn. **57**, 726 (1988).
- [19] K. Machida and M. Kato, Jpn. J. Appl. Phys. **26**, L660 (1987).
- [20] R.H. Heffner *et al.*, Phys. Rev. Lett. **65**, 2816 (1990); G.M. Luke *et al.*, Phys. Rev. Lett. **71**, 1466 (1993).
- [21] G.M. Luke *et al.*, Nature (London) **394**, 558 (1998).
- [22] Y.J. Uemura *et al.*, Phys. Rev. Lett. **62**, 2317 (1989).
- [23] E.H. Brandt, Phys. Rev. B **68**, 054506 (2003).
- [24] Using the same analysis procedure on the data for the $x = 0.1$ sample of Ref. [16] yields $\lambda_{ab} = 254(2)$ nm and $\kappa = 89(10)$ [Fig. 4(a)], fully consistent with the λ_{ab} estimate of these authors; however, our analysis shows that the field dependence of the linewidth is well described by a standard Ginzburg-Landau model (Ref. [23]).
- [25] Because of the more limited field range of the Sm data, the κ estimate for La was used in the Sm analysis.
- [26] R. Khasanov *et al.*, arXiv:0805.1923.